

Moment scaling at the sol - gel transition

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Abstract

Two standard models of sol-gel transition are revisited here from the point of view of their fluctuations in various moments of both the mass-distribution and the gel-mass. Bond-percolation model is an at-equilibrium system and undergoes a static second-order phase transition, while Monte-Carlo Smoluchowski model is an off-equilibrium one and shows a dynamical critical phenomenon. We show that the macroscopic quantities can be splitted into the three classes with different scaling properties of their fluctuations, depending on wheather they correspond to : (i) non-critical quantities, (ii) critical quantities or to (iii) an order parameter. All these three scaling properties correspond to a single form : $\langle M \rangle^\delta P(M) = \Phi((M - \langle M \rangle) / \langle M \rangle^\delta)$, with the values of δ respectively : = 1/2 (regime (i)), $\neq 1/2$ and 1 (regime (ii)), and = 1 (regime (iii)). These new scalings are very robust and, in particular, they do not depend on the precise form of an Hamiltonian.

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I. REVERSIBLE AND IRREVERSIBLE AGGREGATION MODELS

Below we deal with a system of monomers (the basic units) which can aggregate to form the connected clusters. A monomer is considered as a cluster of mass 1 (this is the mass unit). Moreover, monodispersity of the monomeric mass as well as the mass conservation during the aggregation is assumed, i.e., the cluster-masses are always integer numbers in this approach.

The basic sol-gel transition is the appearance at a finite time of an infinite cluster, called *the gel*. 'Infinite' means here that a finite fraction of the total mass of the system belongs to the gel. Note that this definition is applicable both to finite as well as to infinite systems, but contextually and historically, the tools used to study this behaviour have been defined somewhat differently in these two cases [1]. For finite systems, the following moments of the number-mass-distribution n_s (i.e. *the number* of clusters of mass s) are introduced :

$$M'_k = \sum_s s^k n_s \quad , \quad (1)$$

where the summation is performed over all clusters except the largest one. The superscript ' in Eq. (1) recalls this constraint on the allowed values of s . Consequently, the mass of gel-fraction is just $N - M'_1$ with :

$$N = \sum_{\text{all } s} s n_s \quad ,$$

the total mass of the system. For infinite systems, the following normalized moments of the concentration-mass-distribution c_s (i.e. *the concentration* of clusters of mass s) are introduced :

$$m'_k = \sum_s s^k c_s \quad , \quad (2)$$

where the summation runs over all s . Generally, the concentrations are normalized in a special way :

$$c_s = \lim_{N \rightarrow \infty} \frac{n_s}{N} \quad ,$$

and not in a direct relation with the volume. Consequently, the probability for a monomer to belong to the gel-cluster is just equal to $1 - m'_1$.

These different definitions of moments are equivalent in the sense that they are connected through the relation :

$$m'_k = \lim_{N \rightarrow \infty} \frac{M'_k}{N} \quad .$$

In this paper, we investigate scaling behaviours of distributions of mass-distribution-moments for the two 'classical' approaches to the sol-gel transition : the reversible model (the percolation [2]) and the irreversible model (the Smoluchowski equations [3]). In the former case, diffusion is unimportant and the reaction between clusters is the relevant step, whereas in the latter case, on the contrary, the reaction between clusters is unimportant

and the diffusion is relevant. In this sense, these two models are believed to belong to the two different classes, and all other models of aggregation are suspected to belong to one of them. For example, statistical Flory-Stockmayer theory [4] should behave as the percolation model for the scaling properties, since it corresponds to the calculation 'at equilibrium'. This '*universality*' in diverse sol-gel situations is at present a guess.

Percolation model [1] can be defined as follows : in a box (a part of the regular lattice), each site corresponds to a monomer and a proportion p of active bonds is set randomly between sites. This results in a distribution of clusters defined as ensemble of sites connected by active bonds. For a definite value of p , say p_{cr} , a giant cluster almost surely spans all the box. For example, in the thermodynamic limit when the size of box becomes infinite (this limit will be denoted by 'lim' in the following), a finite fraction of the total number of vertices belongs to this cluster. Therefore, we get the results : $m'_1 = 1$ for $p < p_{cr}$ and $m'_1 < 1$ for $p > p_{cr}$. Moreover, m'_1 is a decreasing function of the occupation probability. This typical behaviour is commonly (and incorrectly) called : '*the failure of mass conservation*', but, as stated before, m'_1 is more simply the probability for a vertex to belong to some finite cluster.

On the other hand, the infinite set of Smoluchowski equations [3] :

$$\frac{dc_s}{dt} = \frac{1}{2} \sum_{i+j=s} K_{i,j} c_i c_j - \sum_j K_{s,j} c_s c_j \quad , \quad (3)$$

are the coupled non-linear differential equations in the variables c_s , i.e., in the concentrations of clusters of mass s . The time t includes both diffusion and reaction times, and these equations suppose irreversibility of aggregation, i.e., the cluster fragmentation is not allowed. The coefficients $K_{i,j}$ represent the probability of aggregation between two clusters of mass i and j per unit of time. Some of them are explicitly known for different experimental conditions [5]. But all such known aggregation kernels have the remarkable homogeneity feature :

$$K_{ai,aj} = a^\lambda K_{i,j} \quad ,$$

for any positive a , with λ called the homogeneity index. Maybe the simplest example of the homogeneous kernel is $K_{i,j} = (ij)^\mu$. It has been shown theoretically that if μ is larger than $1/2$, then there exists a finite time, say t_{cr} , for which m'_1 becomes smaller than 1 for $t > t_{cr}$ [6]. This can be interpreted as the appearance of an infinite cluster at the finite time t_{cr} , and it is tempting to put in parallel the occupation probability p in the percolation model and the time t in the Smoluchowski approach. But we will see later that even if this parallel seems reasonable (p being the advancement of the aggregation process), some physical quantities behave quite differently. Note at last that in eq. (3), as written above, the sum over j does not include the gel $j = \infty$ if any (since $c_\infty = 1/\infty = 0$), so the reaction between sol and gel is not taken into account. In principle, an additional term realizing the sol-gel aggregation should be added [7] on the right-hand member of eq. (3).

II. REVERSIBLE AND IRREVERSIBLE SOL-GEL TRANSITIONS

To see differences between both types of models, we shall treat two particularly simple cases : the bond-percolation on the Bethe-lattice, and the aggregation kernel $K_{i,j} = ij$ in

the Smoluchowski approach. They are chosen to be as close as possible in the sense that clusters generated by the percolation model are branched structures (without any loop) in an infinite-dimensional space. So, all their constituents are at the surface and reactivity must then be proportional to their mass [7]. If the diffusion of clusters is negligible, e.g., when all clusters diffuse with the same velocity, or for highly concentrated systems, the corresponding reactivity kernels $K_{i,j}$ between two such clusters of mass i and j should be proportional to ij .

The bond-percolation on the Bethe-lattice with coordination number z , has been solved by Fisher and Essam [8]. Here, the main result we are interested in, is the average concentration [8] :

$$c_s = z \frac{((z-1)s)!}{((z-2)s+2)!s!} p^{s-1} (1-p)^{(z-2)s+z} , \quad (4)$$

and the first normalized moment :

$$m'_1 = \left(\frac{1-p}{1-p^*} \right)^{2z-2} , \quad (5)$$

with p^* being the smallest solution of equation :

$$p^*(1-p^*)^{z-2} = p(1-p)^{z-2} . \quad (6)$$

Let us define $p_{cr} \equiv 1/(z-1)$. For $p < p_{cr}$, the only solution of the above equation is : $p^* = p$, but when p is larger than p_{cr} , then there is a smaller non-trivial solution which behaves as $p_{cr} - |p - p_{cr}|$ near p_{cr} . Above this threshold, the moment m'_1 is smaller than 1 and behaves approximately as $1 - 2(p - p_{cr})/(1 - p_{cr})$. The marginal case $z = 2$ corresponds to the linear-chain case.

Coming back to the concentrations, we can see that for large values of the size s , the following Stirling approximation holds :

$$c_s \sim s^{-5/2} \exp(-\alpha s) , \quad (7)$$

with α given by :

$$\alpha = \ln \left(\frac{p}{p_{cr}} \left(\frac{1-p}{1-p_{cr}} \right)^{z-2} \right) . \quad (8)$$

For this model, a power-law behaviour of the concentrations is seen at the threshold p_{cr} , where more precisely $c_s \sim s^{-\tau}$ with $\tau = 5/2$. Outside of this threshold, an exponential cut-off is always present [9]. This sort of critical behaviour at equilibrium is analogous to the thermal critical phenomena, and in particular, there exist two independent critical exponents, for example τ and σ which is the exponent of the mean cluster-size divergence (here $\tau = 5/2$ and $\sigma = 1$), to describe completely the critical features.

The case of the Smoluchowski equations is quite different. Putting $K_{i,j} = ij$ in these equations, Leyvraz and Tschudi [6] showed that there exists a critical value of the time, say t_{cr} (here : $t_{cr} = 1$), such that the solution is :

$$c_s = \frac{s^{s-2}}{s!} t^{s-1} \exp(-st) \quad \text{for } t < t_{cr}$$

$$c_s = \frac{s^{s-2}}{s!} \exp(-s)/t \quad \text{for } t > t_{cr} \quad , \quad (9)$$

for size-distribution and :

$$m'_1 = 1 \quad \text{for } t < t_{cr}$$

$$m'_1 = 1/t \quad \text{for } t > t_{cr} \quad , \quad (10)$$

for the first normalized moment.

As explained in the previous section, the behaviour of m'_1 characterizes the sol-gel transition, but some other features are interesting to compare to the percolation model. Firstly, one can see that the power-law behaviour is present for $t > t_{cr}$ and *not only at the threshold*, since for large s , we have :

$$c_s \sim s^{-5/2} \exp(-\alpha s) \quad \text{for } t < t_{cr}$$

$$c_s \sim s^{-5/2}/t \quad \text{for } t > t_{cr} \quad , \quad (11)$$

with $\alpha = t - t_{cr} - \ln(t/t_{cr})$. The whole distribution for the finite-size clusters evolves similarly and the appearance of a power-law behaviour is not a sign of the transition but rather a characteristics of the gelation phase. Secondly, it has been proved that for more general homogeneous kernels : $K_{i,j} = (ij)^\mu$, there exists a relation between the exponent τ of the power-law behaviour and the exponent σ of the divergence of the mean size, more precisely : $\tau = \sigma + 2$. Here for $\mu = 1$ we have $\tau = 5/2$ and $\sigma = 1/2$. So, just one exponent is needed to describe the complete critical behaviour. In this sense, the reversible and irreversible sol-gel transitions, though close, are not equivalent.

III. THE ORIGIN OF FLUCTUATIONS IN REVERSIBLE AND IRREVERSIBLE SOL-GEL MODELS

As noticed by Einstein [10], fluctuations of a macroscopic variable M at the equilibrium resembles a Brownian motion in the space of this variable and, hence, one expects the fluctuations to verify :

$$\frac{\langle (M - \langle M \rangle)^2 \rangle}{\langle M \rangle} \sim \text{Constant} \quad . \quad (12)$$

'Constant' means here : independent of the mass of the system. This approach should be true for any short-ranged correlations, i.e., far from any critical behaviour. When close to the critical point, fluctuations are correlated throughout the whole system and Ornstein - Zernike argument yields [11] :

$$\langle (M - \langle M \rangle)^2 \rangle \sim \epsilon^{\nu(d+\eta-2)} \quad , \quad (13)$$

where ϵ is the distance to the critical point, d is the dimensionality of the system, and ν , η are the two critical exponents related respectively to the divergence of the correlation length and to the divergence of the correlation function. In the same way, the average value of M behaves as $\langle M \rangle \sim \epsilon^\beta$, so that, without making any additional assumption about relations between critical exponents, one obtains :

$$\frac{\langle (M - \langle M \rangle)^2 \rangle}{\langle M \rangle^\delta} \sim \text{Constant} , \quad (14)$$

if and only if $\delta = \nu(d + \eta - 2)/\beta$. δ is here a free parameter and should not be confused with yet another critical exponent. 'Constant' in (14) means : independent of ϵ for an infinite system. On the other hand, but by trivial finite-size scaling, this must also be a constant independent of the mass N of the system at the critical point. Moreover, if the standard relations between critical exponents hold, we obtain : $\delta = 2$ for any dimension d (for the Landau-Ginzburg theory we have to replace d by the upper critical dimension $d_c = 4$, above which the mean-field is valid). So the fluctuations of an order parameter in the thermodynamic systems are expected to behave differently at the critical point and outside of it.

The case of irreversible aggregation models is quite different. Fluctuations in off-equilibrium physical processes are hard to analyze because they develop dynamically and, moreover, they often keep memory of a history of the process. In this sense, large fluctuations in such processes cannot be an unambiguous signature of the critical behaviour. In theoretical studies, it is often assumed that the fluctuations are irrelevant for the correct description of the mean properties of the system (e.g. in the Smoluchowski approach). This point will be revisited below.

We will focus on the cluster-mass distribution n_s , the cluster-multiplicity distribution $P(M'_0)$, and the distribution of the gel-mass for finite systems. The cluster-multiplicity M'_0 is just the total number of clusters minus 1, i.e., the largest cluster is omitted. We wish to answer the following questions : (i) what is the importance of the power-law behaviour of the mass-distribution in detecting the critical behaviour? and, (ii) what are the scaling properties of multiplicity distribution and gel-mass distribution near the sol-gel transition for reversible and irreversible models? The importance of the pertinent quantity M'_0 in this context is that it is directly observable in many experimental situations where the system is not too large and the cluster masses are not directly accessible like, e.g., in the hadronization process in strong interaction physics or in the process of formation of the large scale structures in the Universe. Moreover, informations about the normalized moments m'_0 or their fluctuations can also be obtained for large systems, such as in the polymerization, the colloid aggregation, or the aerosol coalescence.

IV. SCALING OF THE MULTIPLICITY-DISTRIBUTIONS AT THE REVERSIBLE AND IRREVERSIBLE SOL-GEL TRANSITION

The multiplicity distribution is intensely studied in the strong interaction physics where simple behaviour of much of the data on hadron-multiplicity distribution seems to point to some universality independent of the particular dynamical process. Some time ago, Koba,

Nielsen and Olesen [12] suggested an asymptotic scaling of this multiplicity probability distribution in strong interaction physics :

$$\langle M'_0 \rangle P(M'_0) = \Phi(z) , \quad z \equiv \frac{M'_0 - \langle M'_0 \rangle}{\langle M'_0 \rangle} , \quad (15)$$

where the asymptotic behaviour is defined as $\langle M'_0 \rangle \rightarrow \infty$, $M'_0 \rightarrow \infty$ for a fixed $M'_0 / \langle M'_0 \rangle$ ratio. $\langle M'_0 \rangle$ is the multiplicity averaged over an ensemble of independent events. The KNO scaling means that data for different energies (hence differing $\langle M'_0 \rangle$) should fall on the same curve when $\langle M'_0 \rangle P(M'_0)$ is plotted against the scaled variable $M'_0 / \langle M'_0 \rangle$. Extending this assumption, we suppose the more general scaling form :

$$\langle M'_0 \rangle^\delta P(M'_0) = \Phi(z_\delta) , \quad z_\delta \equiv \frac{M'_0 - \langle M'_0 \rangle}{\langle M'_0 \rangle^\delta} , \quad (16)$$

with δ a real parameter and Φ a positive function. This form will be called the δ -scaling. KNO-case corresponds to $\delta = 1$. The normalization of the probability distribution $P(M'_0)$ and definition of the average value of M'_0 , provides the two constraints :

$$\begin{aligned} \lim_{-\langle M'_0 \rangle^{1-\delta}}^{\infty} \Phi(u) du &= 1 , \\ \lim_{-\langle M'_0 \rangle^{1-\delta}}^{\infty} u \Phi(u) du &= 0 , \end{aligned} \quad (17)$$

which imply : $\delta \leq 1$ since Φ is positive.

As shown by Botet et al [13], the multiplicity distribution for the 3d-bond percolation model on the cubic lattice at the infinite-network percolation threshold exhibits the δ -scaling with $\delta = 1/2$. Even though the system experiences the second-order critical phenomenon, fluctuations of the multiplicity-distribution remain small and the KNO-scaling does not hold. Of course, m'_0 is *not* in this case an order parameter since $\tau > 2$ even though there is some irregularity in its behavior passing the threshold. This non-analyticity can be illustrated by the exact result for bond-percolation on the Bethe lattice. In this mean-field case, the normalized 0^{th} -moment is :

$$m'_0 = (1 - \frac{z}{2} p^*) \left(\frac{1-p}{1-p^*} \right)^{2z-2} \simeq \frac{z-2}{2(z-1)} - (z-1)\epsilon + (1 - \frac{z}{2})|\epsilon| , \quad (18)$$

with : $\epsilon = p - p_{cr}$, and $\epsilon \ll 1$. It is easy to see that there is a jump of the first p -derivative of m'_0 : $-z/2$ for $p \rightarrow p_{cr}^-$, and $(4-3z)/2$ for $p \rightarrow p_{cr}^+$. The proper order parameter for this model is the normalized mass of the gel-phase, i.e., the mass of the largest cluster divided by the total mass of the system S_{max}/N . Different probability distributions $P(S_{max}/N)$ for different system sizes N can be all compressed into a unique characteristic function (see Fig. 1) :

$$\langle S_{max}/N \rangle P(S_{max}/N) = \Omega \left(\frac{S_{max} - \langle S_{max} \rangle}{\langle S_{max} \rangle} \right) , \quad (19)$$

which is an analogue of the the KNO-scaling function (15) of the multiplicity probability distributions. This new result is important since it seems to be a characteristic critical

behaviour of the order parameter. Now we will discuss what happens for a dynamical transition.

We have simulated Smoluchowski approach by a standard Monte-Carlo binary aggregation [14]. At each step of the event-cascade, a couple of cluster, with masses i and j , is chosen randomly with probability proportional to $(ij)^\mu$, the time is then increased by the inverse of this probability, and the couple of clusters is replaced by a unique cluster of mass $i + j$. The largest cluster is always taken into account in this scenario. Let us discuss here the ($\mu = 1$)-case ($K_{i,j} = ij$). With the proper normalization, the critical gelation time is $t_{cr} = 1/N$, where N is the total mass of the system. The power-law size distribution is indeed recovered numerically with the right exponent $\tau = 5/2$ (see Fig. 2). In contrast to the percolation model, the multiplicity distribution shows ($\delta = 0.2$)-scaling (see Fig. 3) corresponding to very small correlated fluctuations. The scaling function, which in this case is asymmetric and sharp, is also quite different from the one for the percolation [13].

From the point of view of the criticality, the moment M'_1 should be a better candidate. The M'_1 -distribution in the z_δ - variable with $\delta = 0.67$ for masses $N = 1024$, $N = 4096$ and $N = 16384$ is shown in Fig. 4. This non-trivial value of δ is here a signature of the transition : it disappears for $t > t_{cr}$ (see Fig. 5). But this is not yet analogous to the KNO-scaling because M'_1 is not exactly the order parameter. The true quantity to use is the reduced average mass of the largest cluster : $S_{max}/N = (N - M'_1)/N = 1 - m'_1$ for which ($\delta = 1$)-scaling holds (see Fig. 6), as suggested by the phase-transition arguments, and in accordance also with the equilibrium percolation model.

The results we have obtained here for the reversible or irreversible sol-gel transition in thermodynamical or dynamical systems are expected to satisfy the following general conjecture :

- The occurrence of ($\delta = 1$) - scaling in the probability distribution $P(M)$ of a certain macroscopic quantity M is the sign of a critical behavior, with M as the order parameter;
- The occurrence of scaling with $1/2 < \delta < 1$ in $P(M)$ is the sign of a critical behavior in the system with M related to but not identical to the order parameter;
- The occurrence of $\delta = 1/2$ - scaling in the distribution $P(M)$ is the sign that the variable M is not singular.

These conjectures has also recently been checked for kinetic fragmentation systems [15], and confort them.

V. CONCLUSIONS

We have studied here the scaling features of certain macroscopic quantities which are relevant for the sol-gel transition, such as the two first moments of the cluster-mass-distribution and the mass of the gel, i.e., the order parameter. We have employed two different models : the percolation model which exhibits the equilibrium phase transition and the off-equilibrium Smoluchowski theory which shows a dynamical phase transition. The phase transition threshold in both cases is associated with the very particular scaling (19) of the

order parameter fluctuations. For Smoluchowski's equations, this scaling is seen uniquely at the critical gelation time t_{cr} and disappears both for $t < t_{cr}$ and $t > t_{cr}$. It is interesting to notice that for $t > t_{cr}$, the cluster-mass-distribution is a power-law but, nevertheless, the order parameter fluctuations exhibit the small amplitude limit of fluctuations, i.e., the Gaussian scaling.

For quantities other than the order parameter, similar scaling laws (see eq. (16)) are present but now with the different value of the parameter δ of the scaling law. We have shown that the scaling : $\langle M \rangle^\delta P(M) = \Phi((M - \langle M \rangle)/\langle M \rangle^\delta)$ for $M, \langle M \rangle \rightarrow \infty$, holds for any macroscopic quantity and the value of δ is : 1/2 for the non-critical quantities, $0 < \delta < 1$ ($\delta \neq 1/2$ and 1) for critical quantities which are related but which are non identical to the order parameter, and 1 for the order parameter, respectively. These new scaling laws could be used in the phenomenological applications either to find the phase transition threshold or to investigate whether the observed quantity is : regular, critical or whether it corresponds to the order parameter in the studied process. For example, in high-energy lepton-lepton collisions, the scaling behaviour with $\delta = 1$ has been reported in the multiplicity distributions of the produced particles for various collision energies [16]. This behaviour has been interpreted as a signature of the asymptotic limit (the Feynman scaling) in the S -matrix behaviour, in spite of the fact that the Feynman scaling has been experimentally disproved by the continuing rise of particle density in the central region. The analysis presented in this work offers an alternative and much more plausible explanation for these findings in terms of the off-equilibrium phase transition. It is indeed challenging to see the *a priori* unexpected relation between such different physical phenomena as the sol-gel transition phenomenon and the particle production in the ultrarelativistic collisions of leptons. It would also be extremely interesting to apply the mathematical tools proposed in this work in the more typical aggregation phenomena as found in

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Figure captions

Fig. 1

The gel-mass distribution for the 3d-bond percolation model on cubic lattices of different sizes : $N = 8^3$ (crosses), $N = 10^3$ (asterisks), $N = 12^3$ (triangles), and for the fixed bond probability $p_{cr} = 0.2488$. Each point is the average over 200000 events.

Fig. 2

Double-logarithmic plot of the average mass-distribution of the Monte-Carlo Smoluchowski simulations with aggregation kernel $K_{i,j} = ij$, for the two different system masses : 1024 (diamonds) and $N = 4096$ (filled circles) , at the critical gelation-time ($t_{cr} = 1/N$).

Fig. 3

The multiplicity distribution for the Monte-Carlo Smoluchowski simulations with aggregation kernel $K_{i,j} = ij$, for the two different system masses : 1024 (diamonds) and $N = 4096$ (filled circles), in the z_δ -variable ($\delta = 0.2$) , at the critical gelation-time. Each point is the average over 10^5 independent events.

Fig. 4

The M'_1 -distribution for the Monte-Carlo Smoluchowski simulations with aggregation kernel $K_{i,j} = ij$, for the two different system masses : 1024 (diamonds) and $N = 4096$ (filled circles), in the z_δ -variable ($\delta = 0.67$) , at the critical gelation-time. Each point is average over 10^5 independent events.

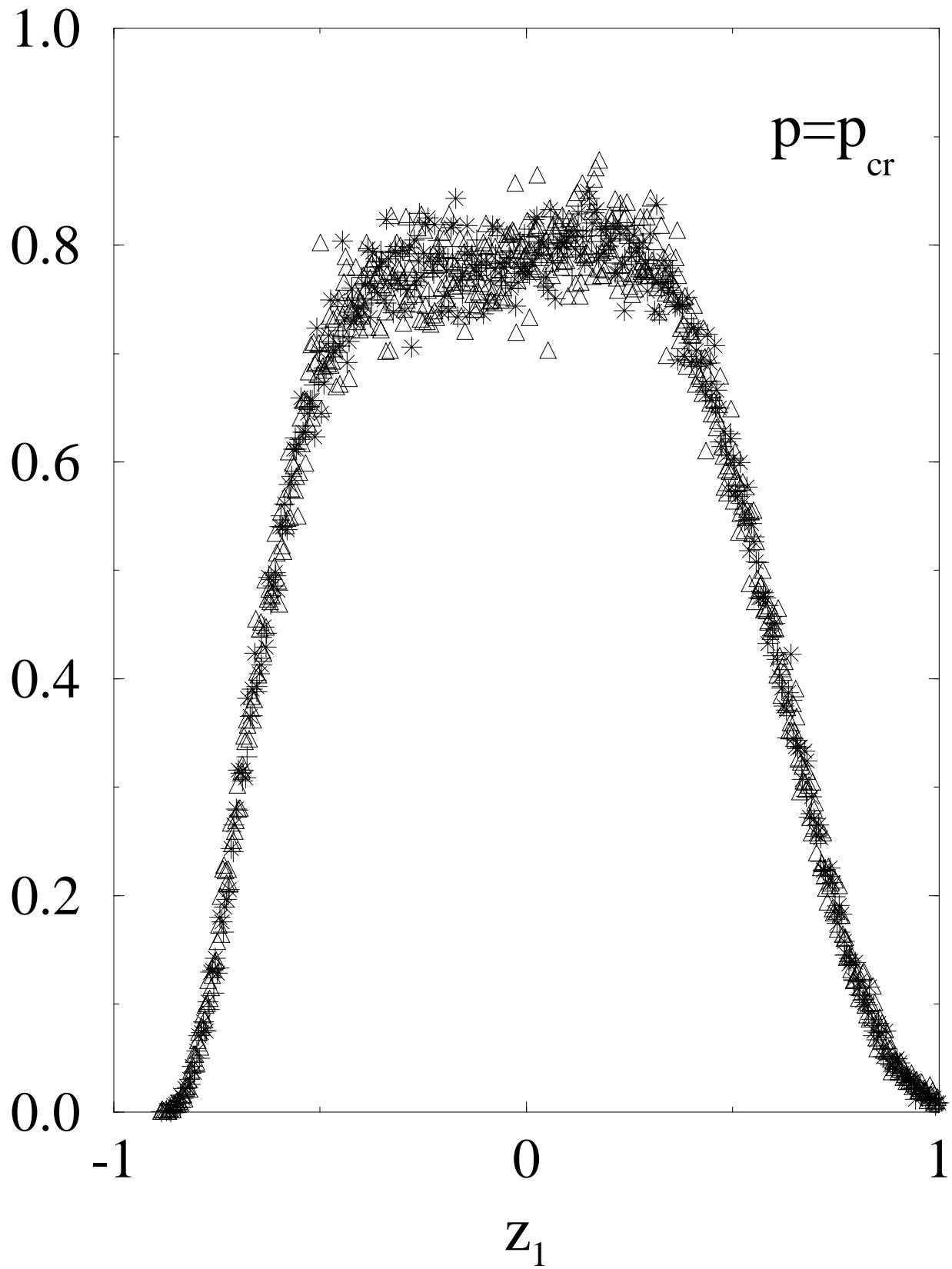
Fig. 5

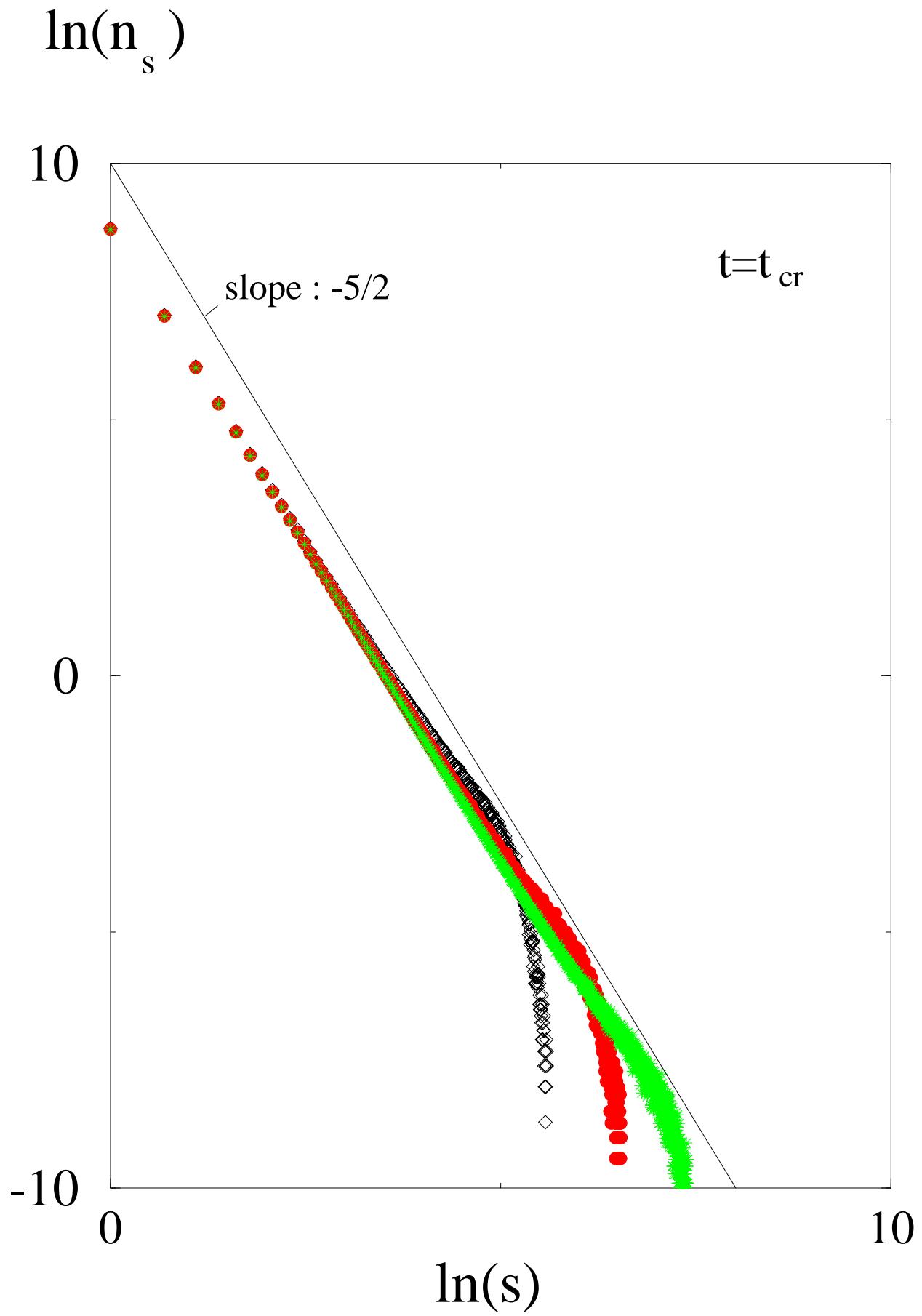
The same as Fig. 4, but in the ($\delta = 1/2$)-variable and after the critical gelation-time ($t_{cr} = 2/N$).

Fig. 6

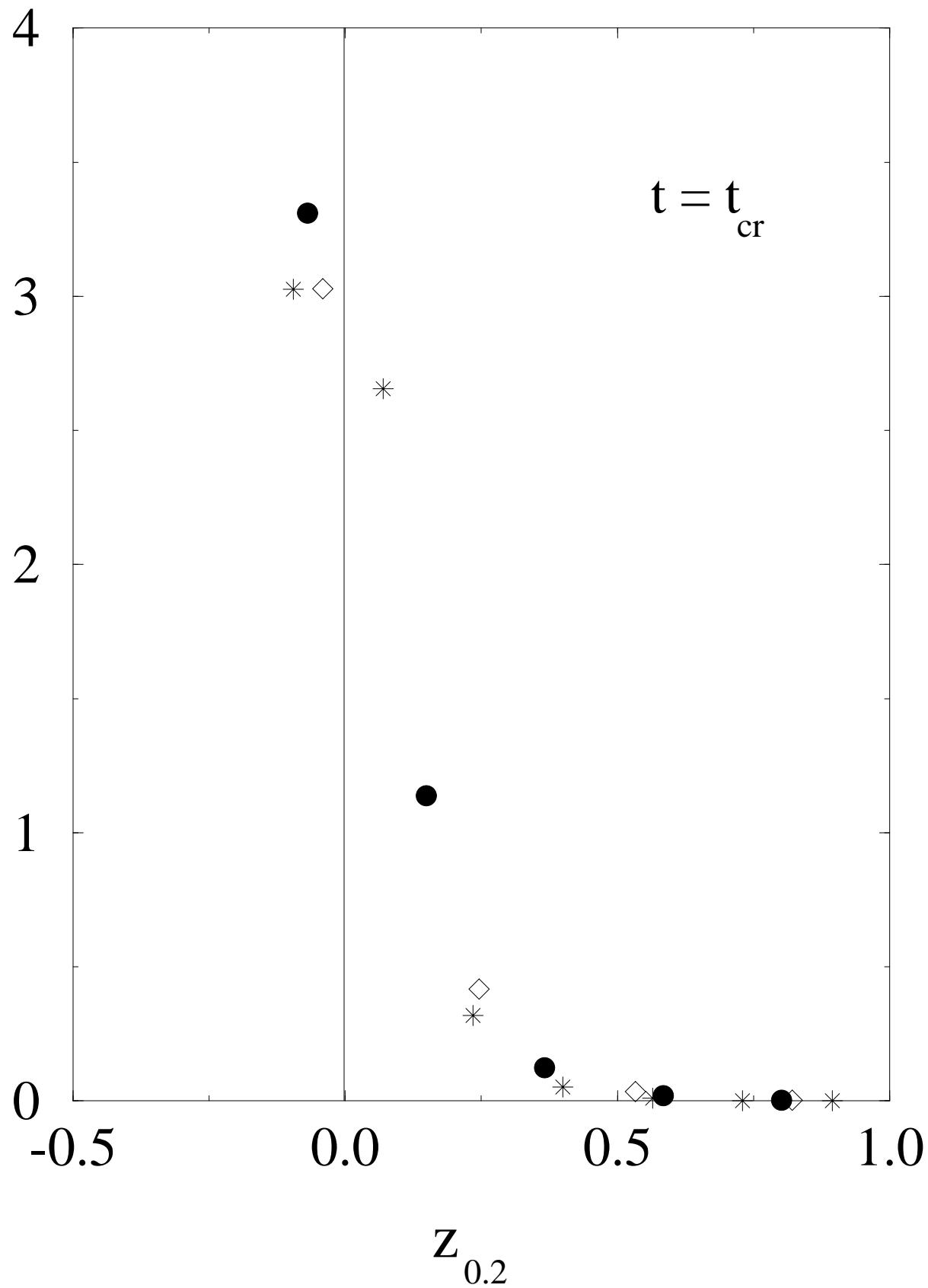
The S_{max} -distribution for the Monte-Carlo Smoluchowski simulations with aggregation kernel $K_{i,j} = ij$, for the two different system masses : 1024 (diamonds) and $N = 4096$ (filled circles), in the KNO-variable at the critical gelation-time. Each point is the average over 250000 independent events.

$$\langle s_{\max} \rangle P(s_{\max})$$

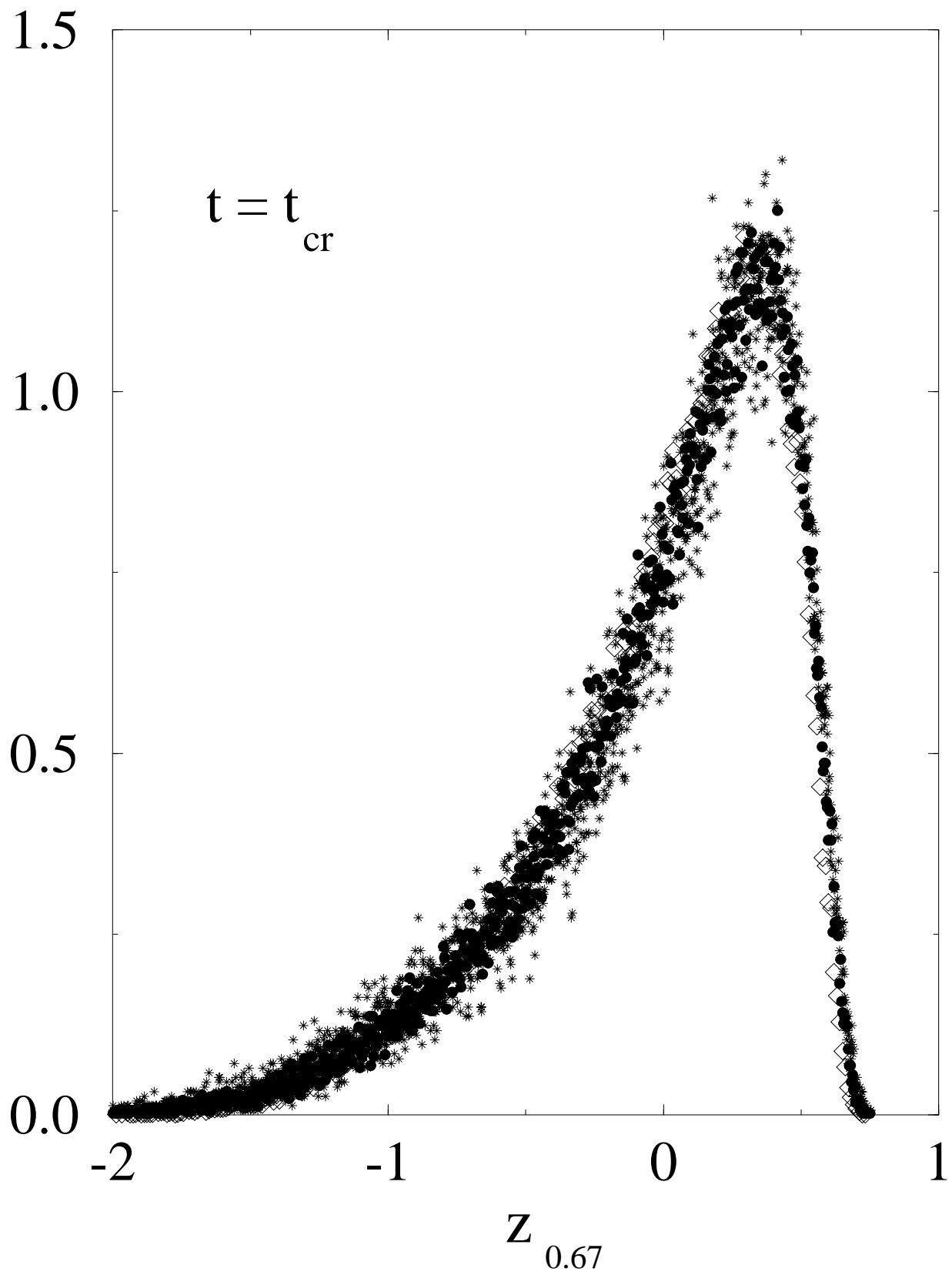




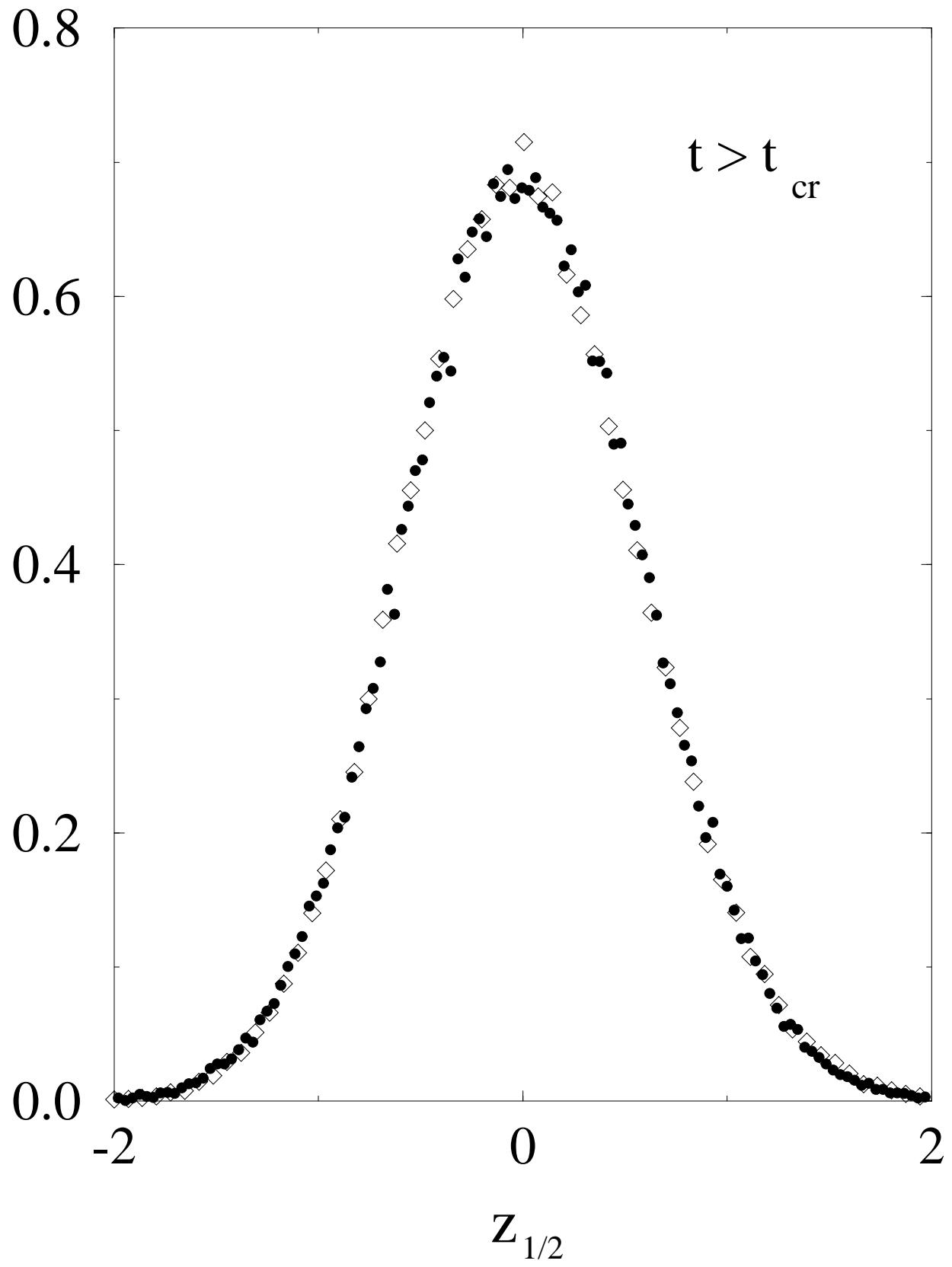
$$\langle M'_0 \rangle^{0.2} P(M'_0)$$



$$\langle M_1^{0.67} \rangle P(M_1)$$



$$\langle M'_1 \rangle^{1/2} P(M'_1)$$



$$\langle S_{\max} \rangle P(S_{\max})$$

